Coherence properties of a broadband femtosecond mid-IR optical parametric oscillator operating at degeneracy

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Abstract: We study coherence properties of a $\chi^{(2)}$ optical parametric oscillator (OPO), which produces 2/3-octave-wide spectrum centered at the subharmonic (3120 nm) of the femtosecond pump laser. Our method consists of interfering the outputs of two identical, but independent OPOs pumped by the same laser. We demonstrate that the two OPOs show stable spatial and temporal interference and are mutually locked in frequency and in phase. By observing a collective heterodyne beat signal between the two OPOs we show that one can deterministically choose, by cavity length adjustment, between the two frequency states corresponding to the two sets of modes shifted with respect to each other by half of the laser pulse repetition rate. Moreover, we observe that the existence of two opposite phase states, a known common feature of a parametrically driven $n = 2$ subharmonic oscillator, reveals itself in our experiment as a common phase, 0 or π, being established through the whole set of some 300 thousand longitudinal modes.

OCIS codes: (190.4975) Parametric processes; (190.4410) Nonlinear optics, parametric processes.

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1. Introduction

Extending the operating range of broadband frequency combs and, more generally, few-cycle carrier-envelope-offset stabilized optical pulses - to the mid-infrared (> 2.5 μm) wavelengths is advantageous for a wide range of applications including X-ray production via high harmonic generation [1], attosecond physics [2], laser-driven particle acceleration [3], broadband spectroscopic trace gas detection [4] and molecular fingerprinting [5]. Several techniques are being developed including direct laser sources [6], optical rectification [7, 8], difference-frequency generation [9–11], optical parametric oscillators [12, 13], and whispering gallery microresonators [14].

Recently, we implemented a new method suitable for generating broadband mid-IR coherent output. The source is based on a degenerate (subharmonic) synchronously pumped optical parametric oscillator (OPO), which rigorously both down-converts and augments the spectrum of a pump frequency comb provided by a commercial 1560-nm mode-locked Er-fiber laser. Low intracavity dispersion, exceptionally large parametric gain bandwidth at degeneracy combined with extensive cross mixing of comb components, resulted in extremely broad instantaneous mid-IR bandwidth of 2.5 to 3.8 μm [15]. Similar approach allowed producing broadband mid-IR output spanning 4.4-5.5 μm from a degenerate GaAs-based OPO pumped by femtosecond Cr:ZnSe laser pulses at 2.45 μm [16].

A question arises whether such a 'frequency divider' OPO system preserves coherence properties of the pump laser. For example, it is very tempting to use a commercial carrier-envelope-phase-stabilized frequency comb as a pump and transpose its coherence to the longer-wavelength broadband output. Prior studies of a near-IR 'divide-by-2' degenerate OPO pumped by a femtosecond Ti:Sapphire laser [17, 18] have shown that the OPO longitudinal modes were phase-locked to the pump, which was confirmed by observing stable interference between different spectral slices of the spectrally broadened pump and corresponding spectral portions of the frequency-doubled OPO output. However distinction between the two frequency states and the two phase states of a subharmonic OPO [18] was lost in this experiment: after frequency doubling the OPO output, these two states both collapse to the same second-harmonic field.

Here we present results of a careful study of coherence properties of a broadband degenerate mid-IR OPO, which we performed by a new method, namely by interfering the outputs of two identical, but independent OPOs pumped by the same ultrafast laser. Our method allowed us to demonstrate for the first time: (i) the existence of two stable and distinct frequency states corresponding to the two sets of modes shifted by half the laser mode spacing and (ii) the existence of two distinct phase states, 0 and π, established over the whole set of longitudinal modes.

2. Theory

Resonant systems with parametric excitation that show subharmonic response are known in acoustics and mechanics since at least 19th century. For example, Faraday observed that the standing waves on liquid surfaces oscillated with one-half the frequency of the applied vertical periodic acceleration [19]. Subharmonic parametric oscillators were later widely explored in the microwave [20] and optical domains [21–24]. The common feature of these devices is that (i) 100% of the driving power can be converted into the subharmonic, (ii) subharmonic oscillation is phase locked to the driving force, and (iii) for a frequency-divide-by-*n* subharmonic oscillator there are *n* stable solutions for the phase of the subharmonic response with respect to the stimulus, which differ from each other by $2\pi/n$ radians. Based on the latter property, electrical circuits containing nonlinear inductance or capacitance were proposed and used as memory and logic circuit elements for early digital computers [25, 26].

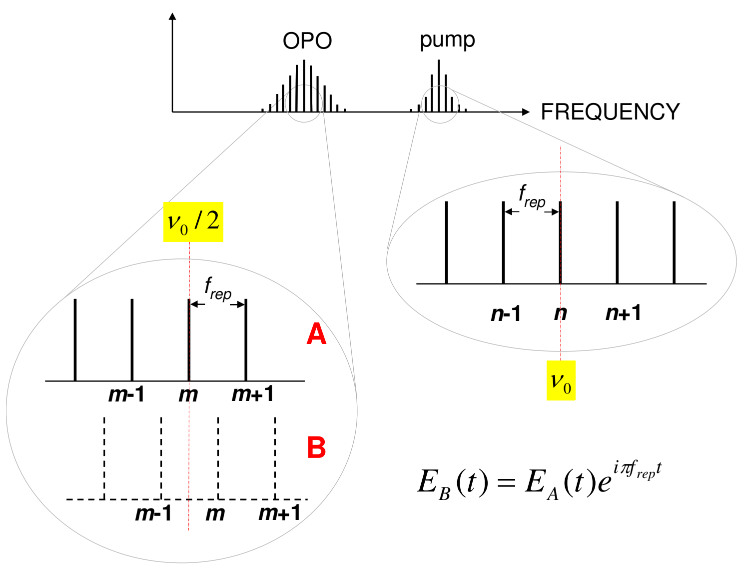

Fig. 1. Modes of a synchronously pumped degenerate OPO. Only two sets of longitudinal modes (A and B) are allowed by photon energy conservation.

For an OPO pumped by a laser at frequency ν_p, the photon energy conservation dictates that $\nu_p = \nu_s + \nu_i$, where ν_s and ν_i are the OPO signal and idler frequencies. Assuming that electric fields of the pump, signal, and idler are correspondingly in the form $E_p\cos(2\pi\nu_p t + \phi_p)$, $E_s\cos(2\pi\nu_s t + \phi_s)$, and $E_i\cos(2\pi\nu_i t + \phi_i)$, the relationship between their phases is expressed as:

$$\varphi_p = \varphi_s + \varphi_i + \pi/2 + \text{integer} \times 2\pi, \quad (1)$$

which ensures the energy flow from the pump to the signal and idler waves [27]. The signal-idler phase difference, $\phi_s - \phi_i$, however is free to adopt any value as long as Eq. (1) is satisfied

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[23, 24]. In a degenerate divide-by-2 OPO, this degree of freedom disappears when the (co-polarized) signal and idler become indistinguishable, which leads to $\phi_s = \phi_i$. Hence, (1) becomes

$$\varphi_p = 2\varphi_{s,i} + \pi / 2 + \text{integer} \times 2\pi, \tag{2}$$

and phase coherence between the OPO and the pump is established (self-phase-locking). Also, when the subharmonic changes its phase by π (flips sign), the total phase difference changes by 2π and (2) still remains valid. This corresponds to the two *phase states* for $n = 2$ subharmonic, mentioned above, namely "0" and "π".

In the case of femtosecond pumping by a mode-locked laser, the pump and the OPO output are represented by a manifold of longitudinal modes sharing the same mode spacing equal to the pump pulse repetition frequency f_{rep}. Figure 1 sketches the spectrum of a degenerate synchronously pumped OPO in which case there is a spectral overlap and cross-coupling between the signal and the idler modes. Since both of them are resonant, their spectral components need to overlap with the OPO cavity modes. Suppose for clarity that one of the pump laser modes is locked to a CW laser with a frequency ν_0 (Fig. 1). From the requirement of the photon energy conservation and matching cavity resonances, it follows that the OPO modes can be aligned in only two ways. *Case A*, one of the OPO modes coincides with $\nu_0/2$. Case B: the frequency $\nu_0/2$ is exactly halfway between the two neighboring modes [20]. As a result, assuming the pump is a frequency comb with its spectral components represented by

$$\nu_n = f_{CEO} + n f_{rep}, \tag{3}$$

where n is an integer centered around some large number $n_0 = 10^6$ - 10^7 and f_{CEO} is the carrier-envelope offset (CEO) frequency ($0 \le f_{\text{CEO}} < f_{\text{rep}}$), the OPO frequency comb is expressed as either:

$$\nu_{m,A} = \frac{f_{CEO}}{2} + m f_{rep}, \tag{4}$$

or

$$\nu_{m,B} = \frac{f_{CEO}}{2} + (m + \frac{1}{2}) f_{rep}, \tag{5}$$

where m is the OPO mode number (centered around some $m_0 \sim n_0/2$).

The two *frequency states* A and B in Eqs. (4-5) correspond to the two sets of modes shifted in frequency by $f_{\text{rep}}/2$. Their time-domain representations $E_{\text{A}}(t)$ and $E_{\text{B}}(t)$ are hence related as

$$E_B(t) = E_A(t) e^{i2\pi \frac{f_{rep}}{2} t} = E_A(t) e^{i\pi f_{rep} t}. \tag{6}$$

Because the pulse duration is much smaller than the period T, the term $exp(i\pi f_{\text{rep}} t)$ is reduced for each consecutive pulse to 1 or −1. That means that the relative sign of $E_{\text{A}}(t)$ and $E_{\text{B}}(t)$ flips every pulse repetition period $T = 1/f_{\text{rep}}$. Here we note that while the OPO *deterministically* operates at either frequency states A or B - one can switch from one state to another by changing the roundtrip cavity length by approximately half of the OPO center wavelength, - the phase states "0" or "π" are *non-deterministic* and depend on zero-point fluctuations of the vacuum field at the OPO build-up.

3. Experiment

The high-finesse ring-cavity OPO (Fig. 2) was synchronously pumped by a femtosecond Er-fiber laser with the central wavelength 1560nm, pulse repetition rate 100 MHz, pulse duration 70 fs, and average power of ~300 mW. The OPO cavity was formed by gold-coated mirrors

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M1 – M5 and a dielectric mirror M6 with high transmission for the pump and high reflectivity at 2.4-4 0.2 µm. The nonlinear gain medium was a 500-µm-long MgO-doped periodically poled lithium niobate crystal at Brewster angle, with a quasi phase matching period of 34.8 µm designed for subharmonic generation at t≈30°C [15]. The doubly resonant condition was achieved by fine-tuning the cavity length with a piezo actuator (PZT) supporting mirror M_3 (Fig. 2). We actively locked the cavity to one of the resonances using 'dither-and-lock' top-of-fringe stabilization. The pump laser cavity length was dithered with low (~1nm) amplitude at ~10kHz; this modulated the OPO output power seen by a mid-IR InAs photodetector with an 1850-nm longpass filter (Fig. 2). Phase-sensitive detection of its output allowed us to generate an error signal, which was supplied to the PZT via a low-pass (<10 Hz) RF filter and a high voltage amplifier such that the cavity length was locked to resonance. This stabilization system could maintain the OPO oscillation for several days by only compensating the low-frequency cavity fluctuations. (It should be noted here that we did not observe effects of passive top-of-the-fringe stabilization, similar to the one of Ref. 24 in a doubly resonant continuous wave OPO, possibly because of much shorter lithium niobate crystal and hence negligible feedback due to photo-thermal or third-order nonlinear effects).

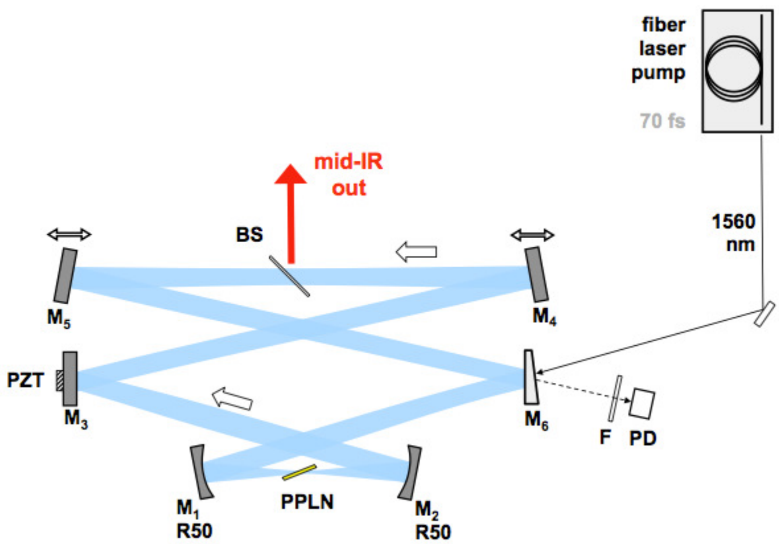

Fig. 2. Ring-cavity degenerate OPO, synchronously pumped by a femtosecond erbium fiber laser. PZT – piezoelectric transducer, BS- pellicle beamsplitter, PD- InAs photodiode, and F – longpass filter.

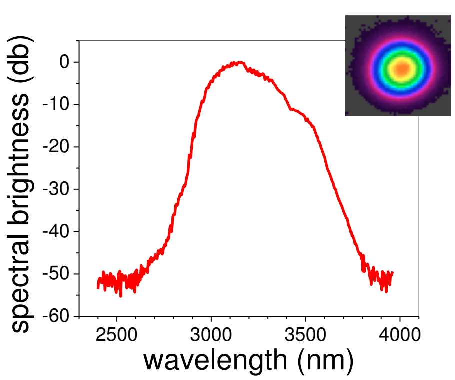

Fig. 3. OPO spectrum measured with a Fourier-transform spectrometer. The inset shows the OPO beam profile.

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Typically, a 2-μm-thick pellicle beamsplitter (8% reflection) was used as beam outcoupler. The OPO pump threshold was 30-100 mW (depending on outcoupling) and at maximum pump the OPO produced 10-50 mW of average power centered at 3120 nm. The spectrum, measured by a Fourier-transform spectrometer (Thermo Nicolet 6700 FT-IR, Fig. 3) spanned from 2500 to 3800 nm at the 20-db level; it corresponds to approximately 2/3 of an octave in frequency and is mainly limited by the intracavity dispersion [20].

To examine coherence properties of our mid-IR comb source, we interfered the outputs of two *identical* subharmonic OPOs (Fig. 4(a)) pumped by the same *free running* femtosecond laser. The beam from the pump fiber laser was split in approximately 50-50% proportion using a half-wave plate and a polarizing beam splitter and each beam was mode-matched to pump OPO_1 and OPO_2, both having a design similar to the one in Fig. 2. The OPO outputs were combined at a small angle using another pellicle beam splitter with 50% reflection and a delay line. The output ports of the beam splitter were directed to a fast InAs detector and a pyroelectric camera (Spiricon, Pyrocam-III). When the delay was adjusted to provide temporal overlap of the pulses, stable interference fringes were observed as a result of interference between the two OPO outputs as in Fig. 4(b), 4(c). These patterns were obtained when both OPOs were operating in the same frequency states *AA* or *BB,* and they were stable for > 10 hours. The existence of stable fringes shows that the generated broadband outputs are frequency- and phase-locked to each other. One can assume that this mutual locking between the two degenerate OPOs happens through locking to the same 'parent' pump laser.

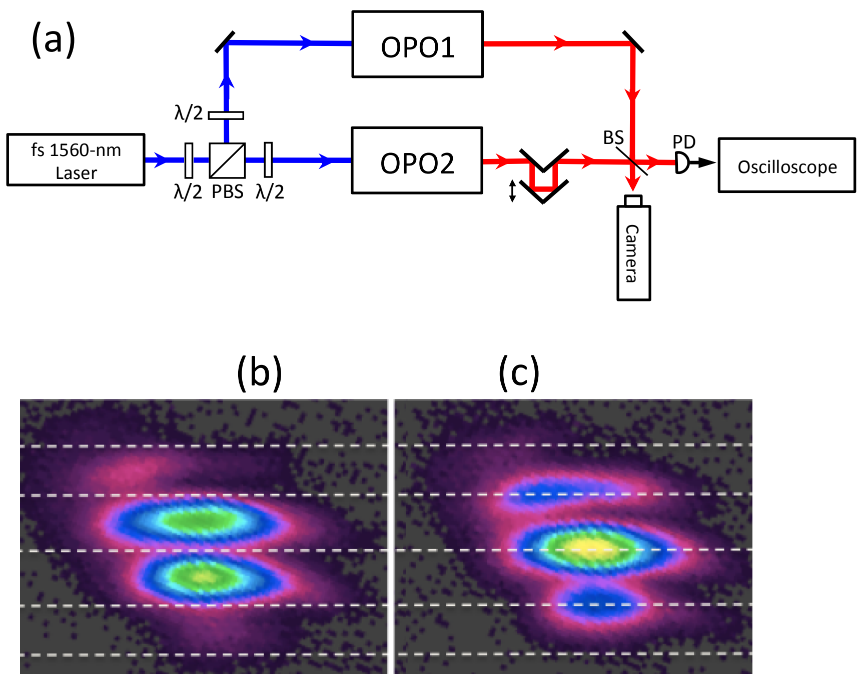

Fig. 4. (a) Schematic diagram of the experimental setup. λ/2 – halfwave plate, PBS – polarizing beamsplitter, BS- pellicle beamsplitter, PD- fast InAs photodiode. (b,c) Complementary interference patterns obtained with an infrared camera. Dashed lines are guides for an eye to show that maximum intensity in (c) corresponds to minimum in (b).

We examined the existence of the two OPO phase states (“0” or “π”) as follows. By blocking and unblocking the cavity path of one of the OPOs, to temporarily quench oscillation, we observed that the fringe pattern randomly switched between two complementary interference patterns of Fig. 4(b) and Fig. 4(c). We interrupted one OPO much faster than the response time of the stabilization system (~0.1 s) so that the cavity length

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and therefore the frequency state were preserved. The existence of two phase states in an optical parametric device has been experimentally verified for a CW degenerate single-frequency OPO [23]. Here we show that a single common phase (randomly chosen between 0 or π) is established across the *whole manifold* of some 300 000 modes; this fact can be accounted for by massive spectral cross-coupling between the OPO modes, similar to establishing phase relations between the laser modes in an actively mode-locked laser.

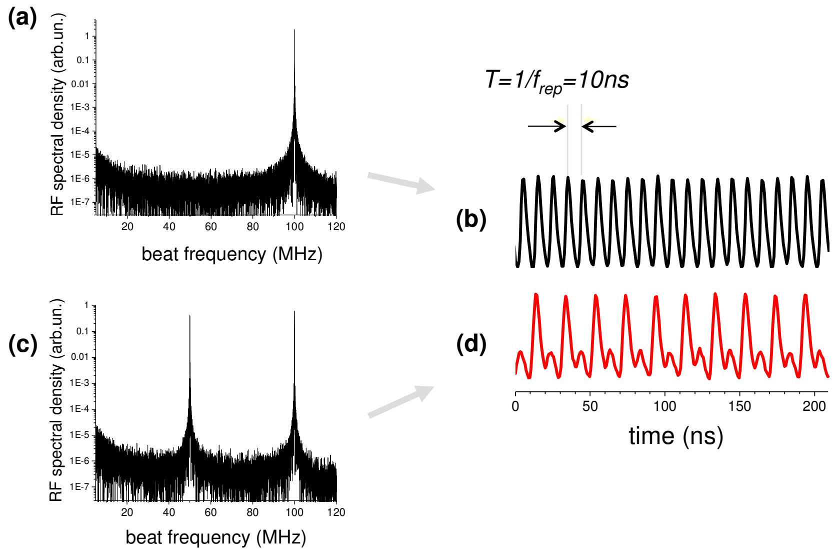

Fig. 5. Time and frequency domain interference between the two identical OPOs. (a) RF spectrogram of the detector signal when both OPOs were in the same (AA or BB) frequency state and (c) in different (AB) frequency states; (b) and (d) are the corresponding time-domain detector signals. 50-MHz ($f_{rep/}2$) beats were observed in the case (c) corresponding to the sequence of pulses separated by 20 ns, which is twice the repetition period of the pump (d).

The time/frequency -domain interference between the outputs of the two OPO was recorded in our experiment by using a fast InAs detector (cutoff wavelength at 3.9 µm). A small detector size, 250 µm, ensured that it detected only a small portion of one interference fringe. Figure 5(a) shows an RF spectrogram (equivalent to a collective heterodyne beat signal) of the detector output recorded in the bright fringe when both OPOs were in the same (*AA* or *BB*) frequency state. The 100 MHz peak on Fig. 5(a) confirms that the frequency comb 'teeth' of both OPOs overlap, while the time-domain detector signal from an oscilloscope of Fig. 5(b) shows a train of pulses separated by $T = 1/f_{rep} = 10$ns. When the cavity length of one of the OPOs was locked to a neighboring resonance, we no longer observed interference fringes. This is the result of the two OPOs being in different frequency states (*AB*) with non-overlapping comb teeth. However a 50-MHz beat-note (Fig. 5(c)) appeared in the RF spectrum; at the same time, the time-domain oscilloscope trace of the detector (Fig. 5(d)) showed a train of pulses with *twice* the repetition period (20 ns) of the pump laser. This can be viewed as an elimination of every other pulse as a result of destructive interference, since the relative sign of the electric fields of the two OPOs is flipped every period *T*, as it follows from (6). In both of the above experiments, we observed that the variation of the frequency comb mode spacing was negligible within the accuracy of our measurements: the frequency peaks in Fig. 5(a), 5(c) were 20-kHz-wide and limited by the acquisition time of 40µs.

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4. Conclusion

We performed a study of coherence properties of a broadband mid-IR degenerate OPO by interfering the outputs of two identical OPOs. We show that their outputs are mutually coherent, which suggests that they are frequency and phase locked to the parent pump laser. We established that such an OPO has two stable frequency states shifted by $f_{rep}/2$ with respect to each other and one can deterministically switch between them. There are also two possible solutions for the phase, differing by π, that are established through he whole comb of modes; one of these solutions is randomly chosen depending on the quantum noise when the OPO starts. Overall, the results obtained suggest that a subharmonic OPO pumped by a carrier-envelope-phase-stabilized frequency comb would inherit all its coherence properties. Future directions for this work include extension of the spectral range to the longer wavelengths, generation of phase-stabilized mid-IR frequency combs and their use in trace molecular detection via coherent Fourier-transform spectroscopy.

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